Supplementary Material for "Phonon transport manipulation in $TiSe_2$ via reversible charge density wave melting"

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I. RESISTIVE CHARACTER OF SCATTERING PROCESSES

As previously noted, the most classical distinction between the resistive character of the processes, both isolated and statistically, is based on the conservation of the quasimomentum. So that, quasi-momentum conserving processes —called Normal processes, $\mathscr{N}$—are considered fully non-resistive (i.e. flux conserving), while the rest—called Umklapp ($\mathscr{U}$)—are deemed as resistive or flux destroyers. However, such a distinction is unphysical and dependent on the choice of the unit cell. Moreover, it fails to describe real materials since there is no real translation between conservation of momentum and that of flux [1]. Indeed, for the former case a more rigorous approach consists in checking whether total velocity between initial and final states is conserved or not. Nevertheless, such a distinction disregards the combined effect of scattering processes as a whole in the flux dynamics; to further illustrate this aspect, consider two scattering events of equal probability such that their combined change in the flux cancels out: though individually resistive, statistically their ensemble is to all effects non-resistive. Therefore, a more rigorous way to account for the resistivity of a material/model consists in employing an *ab initio* linearized collision operator (A), since this naturally gauges both individual and ensemble levels. Mathematically, the collision operator acts on the equilibrium deviations as:

$$\frac{\partial n_i^d}{\partial t}\bigg|_{\text{collision}} = A_{ij} n_j^d, \tag{1}$$

where $\frac{\partial n_i^d}{\partial t}\big|_{\text{collision}}$ is the change in the i-th mode due to collisions/interactions with perturbing fields/elements, and n_i^d is the deviational phonon distribution of the i-th phonon mode from a given equilibrium (Bose-Einstein) distribution. For a detailed description of the terms building the linearized collision operator, A, we refer the reader to Eqs. 3 and 12 in Refs. [2] and [3], respectively. Therefore, the change in the flux due to the collision operator can be obtained as:

$$\frac{\partial \mathbf{J}_i^d}{\partial t}\bigg|_{\text{collision}} = \frac{\hbar\omega_i}{N_{\text{q}} V_{\text{uc}}} \mathbf{v}_i (A_{ij} n_j^d); \tag{2}$$

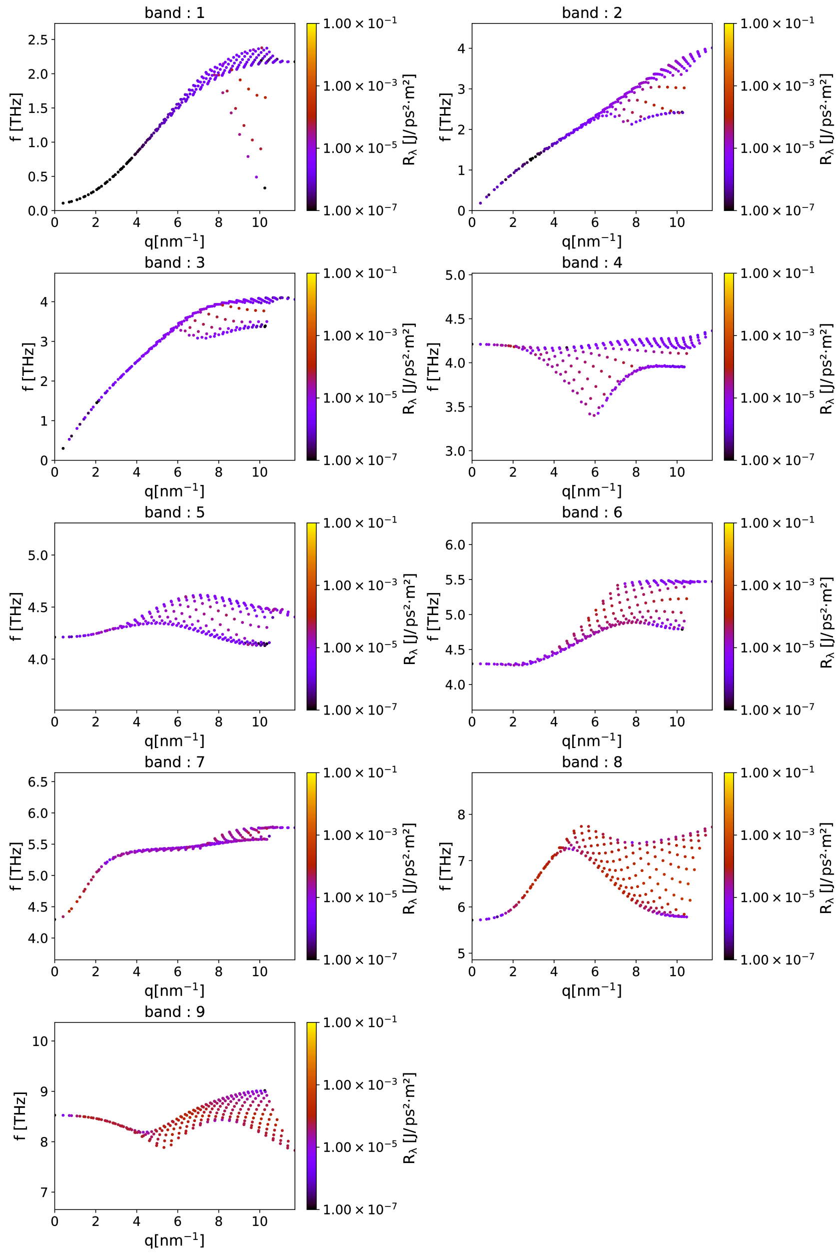
here $\hbar$ is the reduced Planck constant, ω_i is the frequency of the i-th mode, N_{q} the number of points of the regular $\mathbf{q}$-mesh, V_{uc} the volume of the unit cell, and $\mathbf{v}_i$ is the group velocity of the i-th mode.

Consequently, the degree of resistivity of a mode (R_i) for a given reference temperature, namely, the degree in which intrinsic scattering can modify the contribution of a given mode to the flux, can be computed as:

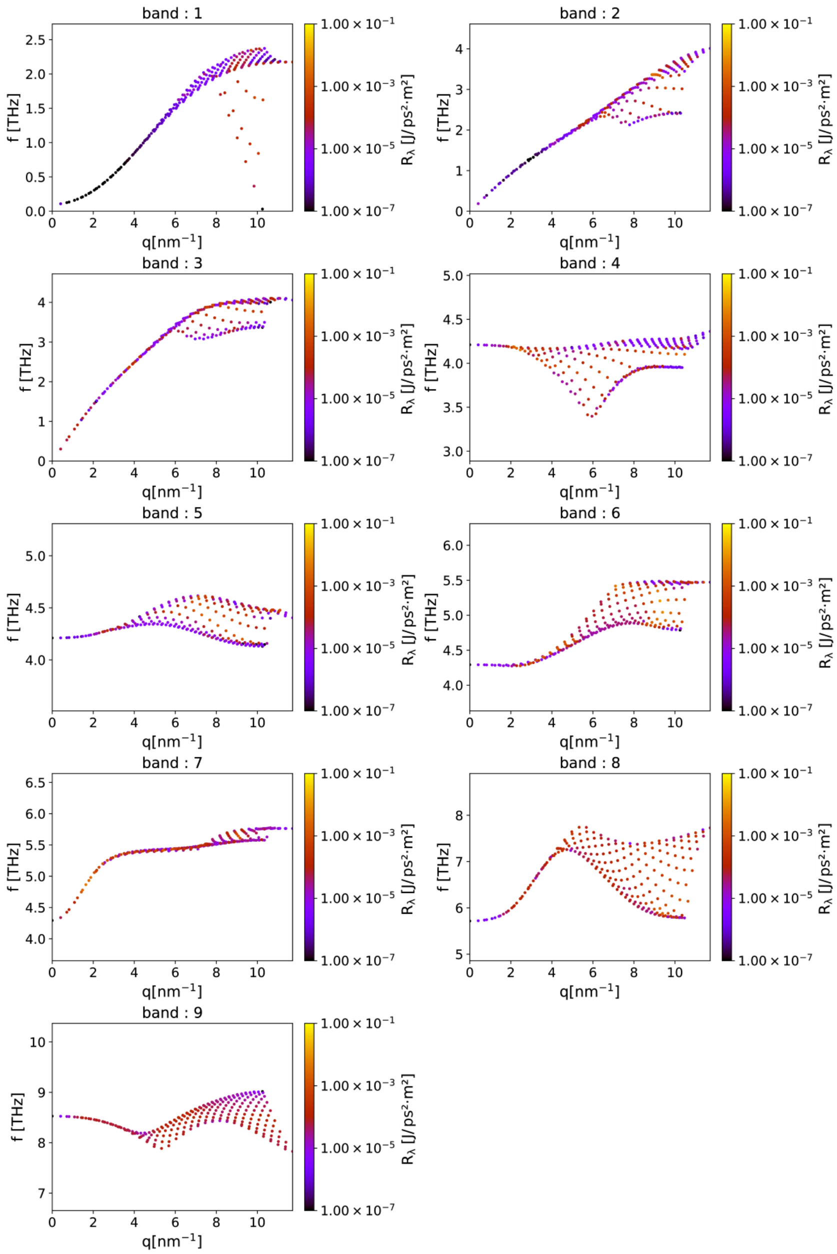
$$R_i = \frac{\hbar\omega_i}{N_{\mathrm{q}} V_{\mathrm{uc}}} \sqrt{\sum_{\alpha} \left[\sum_{j} \mathbf{v}_{j\alpha} \circ (A\mathbf{e}_i)_j \right]^2} \tag{3}$$

where α stands for the Cartesian axis, and $\mathbf{e}_i$ is the i-th canonical basis vector.

The results obtained from this analysis are shown in Figure I and I (see also the main text).

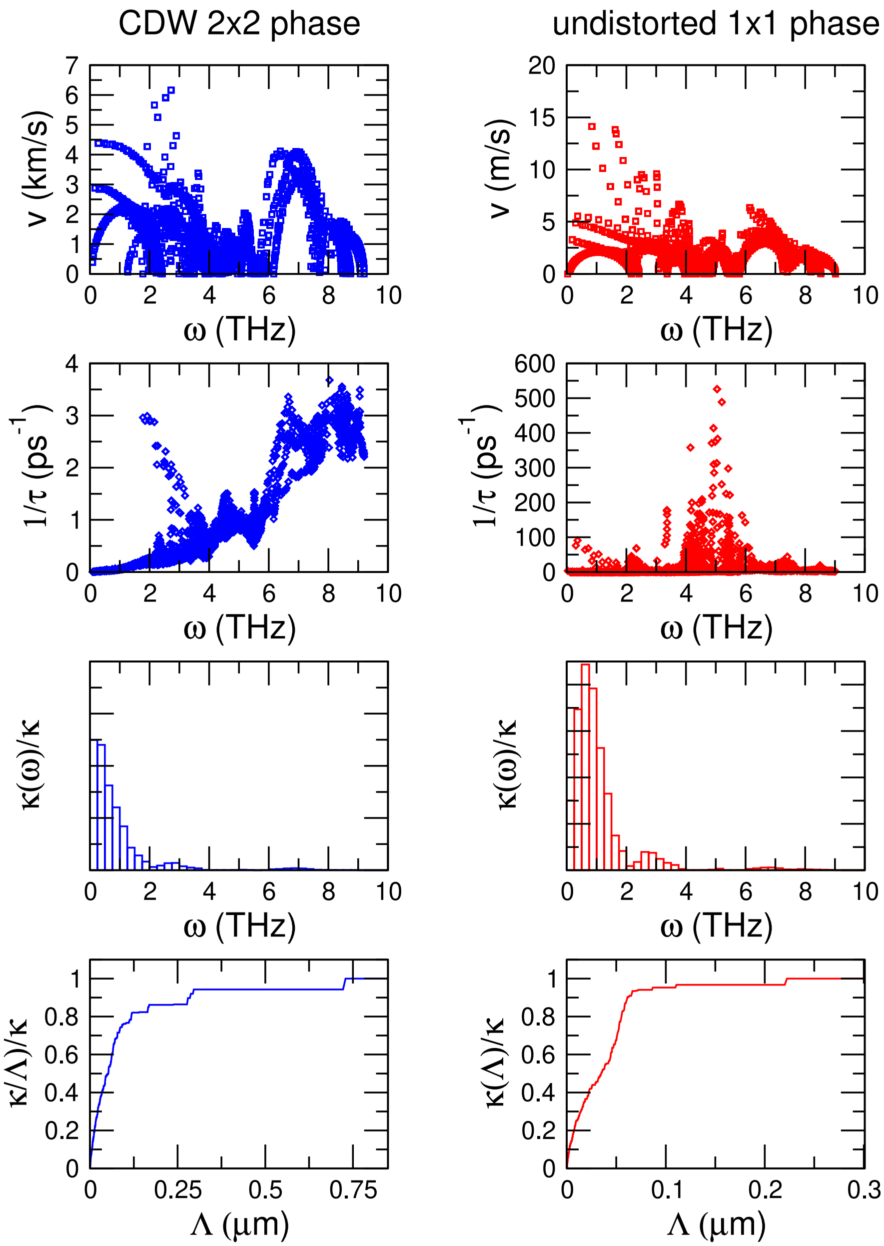


Supplementary Figure 1: Resistive character of phonon-phonon scattering processes for each phonon mode in 1×1 $TiSe_2$ for a high photoexcited charge of $n_e = 0.074640$ e/Å^{-3}.



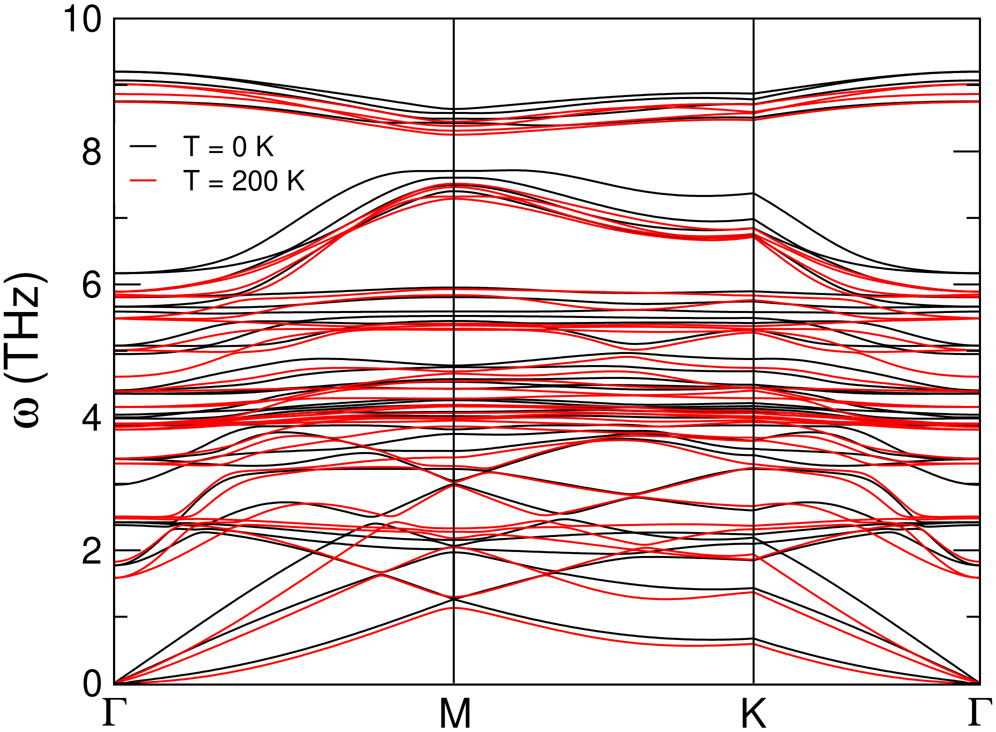
Supplementary Figure 2: Resistive character of phonon-phonon scattering processes for each phonon mode in 1×1 $TiSe_2$ for a low photoexcited charge of $n_e = 0.074378$ e/Å^{-3}.

II. PHONON VELOCITIES, SCATTERING RATES, FREQUENCY RESOLVED κ, AND MEAN FREE PATH CUMULATIVE κ



Supplementary Figure 3: Top to bottom: phonon velocities, phonon-phonon scattering rates, frequency resolved thermal conductivity, and mean free path cumulative thermal conductivity. The frequency resolved κ has been averaged over intervals of 0.25 THz. These plots have been obtained at the RTA level of theory and thus, as discussed in the main text, only provide qualitative trends.

III. T-RENORMALIZED PHONONS OF THE 2×2 CDW PHASE



Supplementary Figure 4: Comparison between zero-kelvin (black) and finite temperature phonon dispersion at 200 K (red) of the 2×2 CDW phase. T-renormalized phonons were obtained with the DynaPhoPy code [4] from *ab initio* molecular dynamics simulations (NVT ensemble, 1.5 fs timestep) and a subsequent normal-mode-decomposition.

[1] A. A. Maznev and O. B. Wright, Demystifying Umklapp vs Normal scattering in lattice thermal conductivity, Am. J. Phys. **82**, 1062–1066 (2014).

[2] Colin D. Landon and Nicolas G. Hadjiconstantinou, Deviational simulation of phonon transport in graphene ribbons with ab initio scattering, J. Appl. Phys. **116**, 163502 (2014).

[3] Martí Raya-Moreno, Xavier Cartoixà, and Jesús Carrete, BTE-Barna: An extension of almaBTE for thermal simulation of devices based on 2D materials, Comput. Phys. Commun. **281**, 108504 (2022).

[4] Abel Carreras, Atsushi Togo, and Isao Tanaka, DynaPhoPy: A code for extracting phonon quasiparticles from molecular dynamics simulations, Comput. Phys. Commun. **221**, 221–234 (2017).